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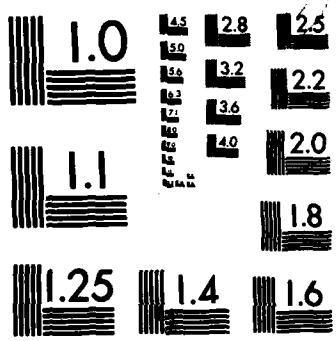

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

An experimental and theoretical study of resonant coherent four wave mixing has been initiated. Prior theoretical investigation by the author has demonstrated that coherent propagation effects can be used to keep all the energy in the radiation field, even in the presence of resonant absorption.

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cont An energy conversion efficiency of 6% was predicted for third harmonic conversion in lithium to 190 nm.

In the period covered by this report (Nov. 1, 1981 - October 31, 1982) we have

(1) Constructed a dye laser amplifier, and achieved an output of 1 mJ for wavelengths covering the most important resonances of lithium (571 nm, 672 nm, 639 nm);

(2) Purchased and assembled the hardware of a data acquisition system, to accurately characterize each pulse (amplitude, duration, phase modulation) and perform the measurement of two photon absorption and third harmonic as a function of the relative phase and delay in the pulse sequence;

(3) Investigated the feasibility of increasing the conversion efficiency through the use of "natural" or "induced" autoionizing resonances; and

(4) Made a theoretical study of the influence of coherence on three photon ionization in lithium.

SCIENTIFIC REPORT

COHERENT PROPAGATION AND SUM FREQUENCY GENERATION INTO THE VACUUM-ULTRAVIOLET

1. OBJECTIVES AND APPROACH

Our broad objective is to implement a scheme of harmonic generation in metal vapors, by which the highest conversion efficiency to the VUV can be obtained. This goal will be reached by exploiting the following properties:

- a two photon resonance, to reduce the peak power needed for maximum conversion;
- coherent propagation to minimize the energy left (lost) in the medium;
- Choice of pulse sequence/frequency/phase to minimize multiphoton ionization.

2. PROGRESS SUMMARY

In the period of performance of this program (Nov. 1, 1981 - October 31, 1982), we have constructed and assembled the main component for the experiment (source and data acquisition equipment). We have investigated the possibility of enhancing the conversion efficiency by the use of natural "induced" photoionization resonances. Numerical and analytical calculations of two photon resonant three photon ionization led to a better understanding of multiphoton resonant ionization, and its dependence on the coherence of the radiation.

3. THEORY

3.1. Background

A two photon resonance is generally known to enhance third (or sum frequency) harmonic generation.[1] However, the stronger the resonance, the lower the maximum efficiency that can be achieved. The advantages in the 2-photon enhancement is to reduce the power at which optimum conversion is achieved, as shown in a theoretical analysis for this project.[2] In general, the maximum conversion efficiency will not be reached because of two-photon absorption. Depletion of the fundamental by two-photon absorption not only reduces the field available for the conversion process, but also affects the resonance condition (intensity dependent through Stark shifting) and the phase matching condition (as the populations of the various levels change with position, so does the contribution of these levels to the susceptibility of the medium).

We have shown theoretically[2,3] that the losses resulting from two photon absorption can be minimized through appropriate pulse shaping, in the conditions of "coherent interaction" (i.e., pulses short compared with the collision times in the metal vapor). Optimum conversion should be achieved with a sequence of two short pulses, with a relative delay of an uneven number of quarter light periods $(2N + 1)T/4$ (of the order of one pulsewidth). With such a pulse sequence, the energy lost by the first pulse to the medium is restituted to the second by two-photon stimulated emission. The peak intensity of the second pulse remains roughly constant over long propagation distances, because the energy transferred from the first pulse compensates for the energy lost to the harmonic field. Even the fraction of ions photoionised is much smaller

than could be expected from considerations only of the energy and duration of the excitation. This is because the atoms spend only a fraction of the irradiation time in the excited state.

3.2 Autoionizing Resonances

As shown by Hodgson et al.[4] and our study[2], the proximity of the resonance at the harmonic frequency will result in enhanced peak conversion efficiency. There are two mechanisms of equal order by which the atom in excited state can return to the ground state: either by emitting two photons at the fundamental frequency (two-photon stimulated emission), or by a two photon process involving a photon at ω_1 and the "harmonic" photon at $2\omega + \omega_1$. The probability ratio of the two processes is a measure of the maximum achievable conversion efficiency. It is also a number characteristic of the particular atomic system being considered. That probability ratio - hence the maximum achievable conversion efficiency - is largest in the proximity of a real resonance at $2\omega + \omega_1$. A "resonance" in the maximum achievable conversion efficiency can thus be found by tuning the frequency ω_1, in order to approach an autoionizing state in the continum with the combination $2\omega + \omega_1$.

3.3. Laser Induced Resonance

This resonance condition (with an autoionizing state) restricts the range of wavelengths over which optimum efficiency can be achieved. Heller and Popov[5] proposed a scheme to induce an autoionizing-like resonance with an external laser field. The level scheme is sketched in Fig. 1. Instead of two two-photon processes connecting the level 2 to the ground state 1, we have two other two photon processes connecting levels 3 to 1 ($\omega_2 - \omega_p$) and levels 3 to 2 ($\omega_1 - \omega_p$) the exact resonance

conditions and intensities of the field will determine whether the field at ω is absorbed or enhanced. For the particular transitions of interest in lithium, we estimate that a laser intensity of the order of a few MW/cm^2 at ω_p will have a significant effect.

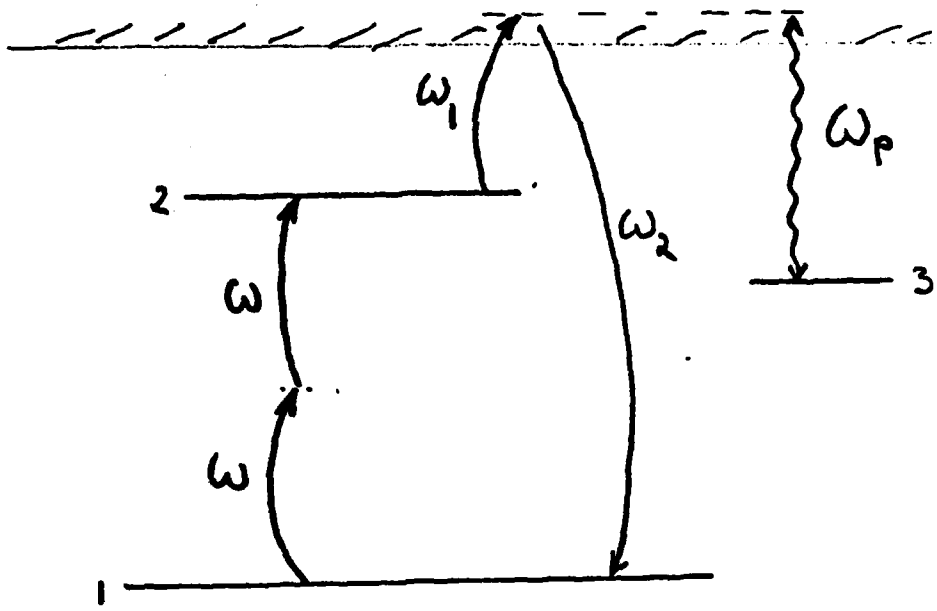
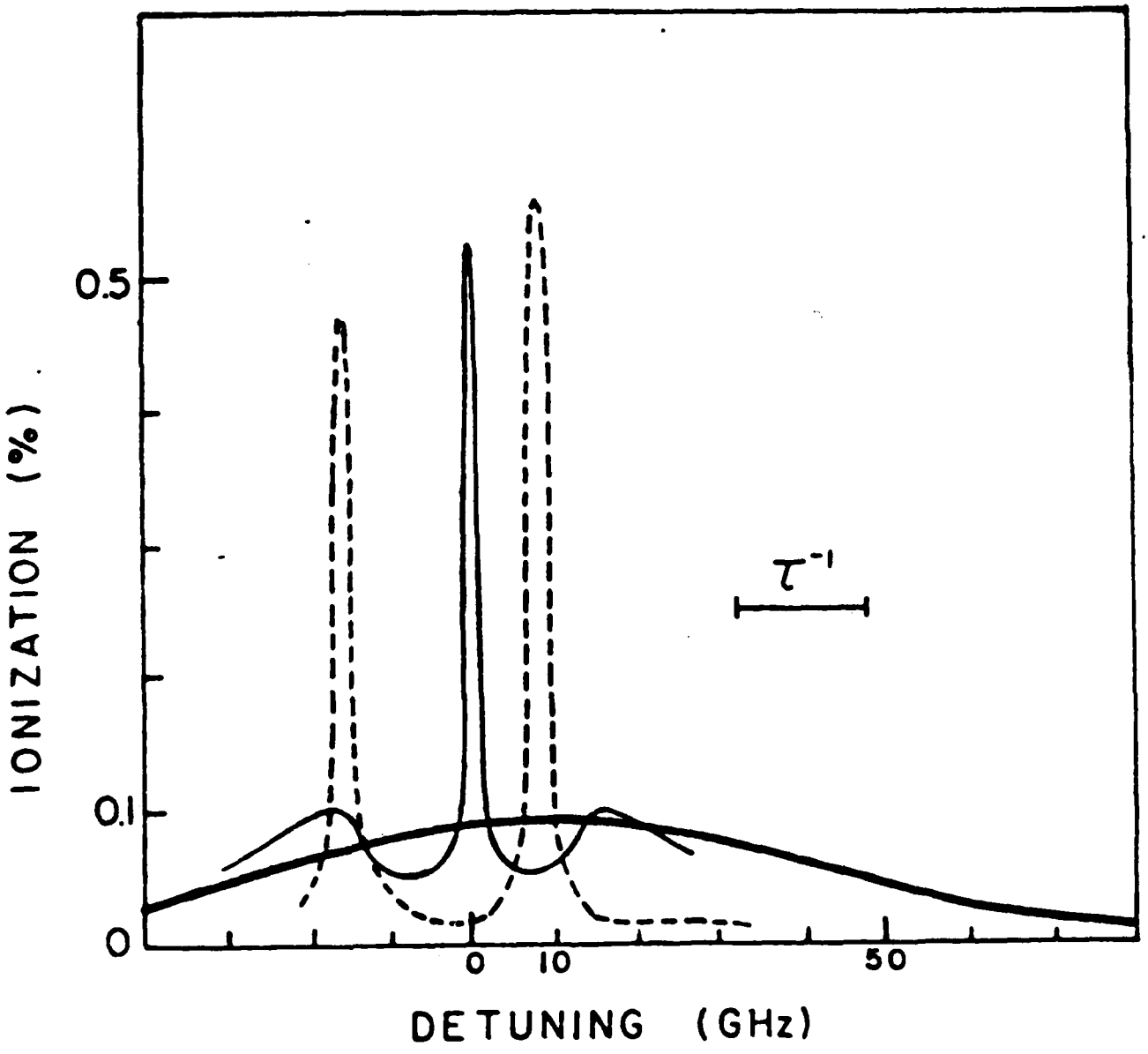


Figure 1

3.4. Influence of Coherence on Multiphoton Ionization.

The sources and pulse shaping devices discussed in the experimental section below give us an ideal opportunity to verify our recent predictions on the influence of coherence on multiphoton ionization [6]. A typical result of the computer modeling is shown in Fig. 2. These plots pertain to three photon ionization, in the presence of a two-photon resonance (2s - 4s transition) in lithium vapor. The excitation consists of a sequence of 10 identical pulses (in shape and frequency). The average number of ions (average over a large number of "experiments" - in general 500) is plotted as a function of the pulse frequency. The thick curve corresponds to incoherent irradiation, modelled by a random distribution of pulses (random in phase and time). The ionization curve is broad and featureless. Nearly an order of magnitude larger number of ions can be produced at a particular frequency if the 10 pulses are exactly in phase (thin solid line). We have instead a deep minimum in the number of ions at a particular frequency if the successive pulses are shifted in phase by 90° (dashed line). This particular type of sequence is also used in our scheme for harmonic generation, in order to minimize the losses due to photoionization.



Some comment should be made on this rather unconventional approach to the study of the influence of source coherence on multi-photon excitation. The dependence of multiphoton ionization on laser coherence was studied a few years ago[7] by varying the number of oscillating laser cavity modes between one and one hundred ("one hundred modes" being considered as a good approximation to incoherent radiation). Such an experiment is not meaningful in presence of various resonances. This

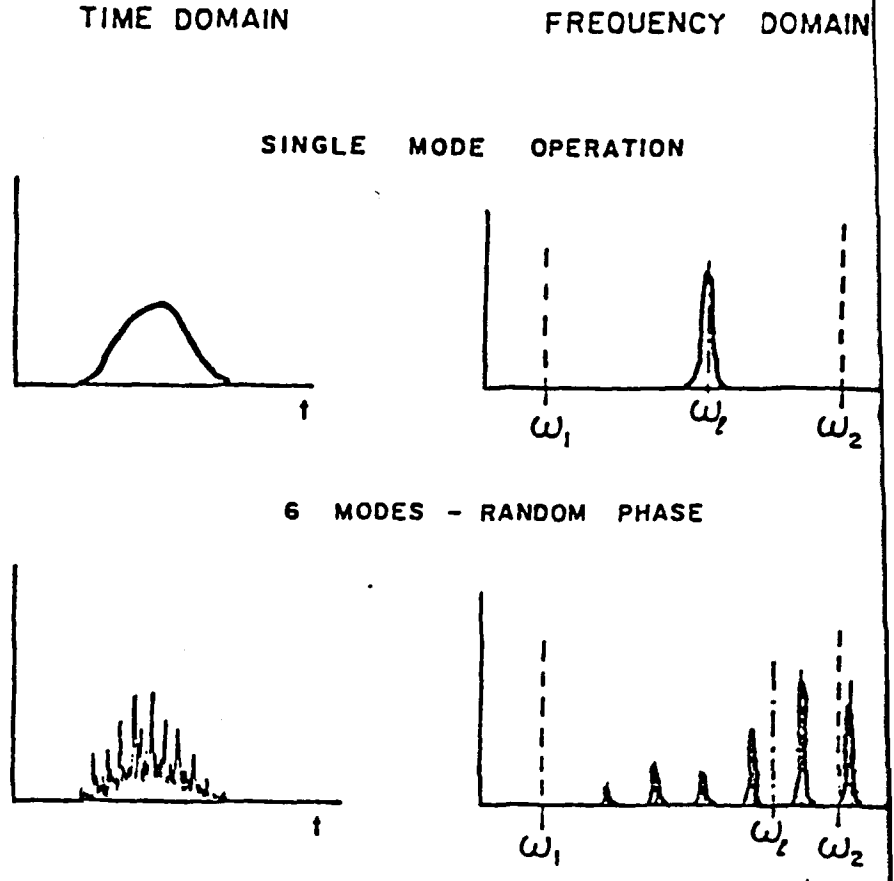


Figure 3.

Changing "coherence" by varying the number of modes. ω_ℓ is the average light frequency. ω_1 and ω_2 are material resonances.

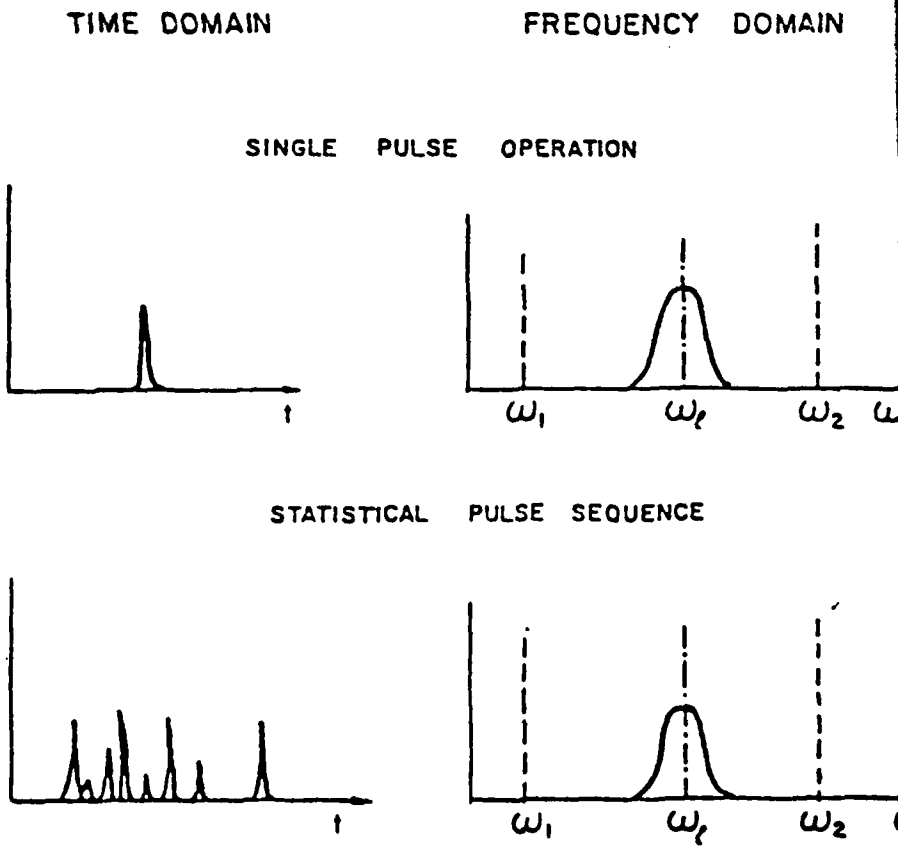


Figure 4.

Changing "coherence" by varying the temporal distribution of picosecond pulses.

point is illustrated in Fig. 3, where ω_ℓ is the average laser frequency, and ω_1 and ω_2 are material resonances. In changing for instance the number of modes from 1 (upper part of the figure) to 6 (lower part), the two resonances have been incorporated in the radiation spectrum. Instead of changing the "parameter coherence", it is the resonance condition that has changed. In this approach, the change in coherence implies a change in spectral width. Instead of studying the influence of coherence (on a multiple photon process) at constant pulse duration (and constant energy density) we propose to study the influence of light coherence at a constant pulse spectral width (and energy density). The proposed situation (Fig. 4) ressembles the case of Fig. 3 where time and frequency domains would have been interchanged. The "coherence" is modified in the time domain by modifying the (temporal) distribution of a pulse sequence (each pulse of the sequence is assumed to have identical shape and frequency). As shown in Fig. 4, the overlap or nonoverlap of the pulse spectrum with material resonances remain unchanged. It is now meaningful to tune the average pulse frequency across the resonances ω_1 and ω_2 and to measure the resonance curves (of multi-photon ionization) for various statistical characteristics of the laser source.

There are many more "statistical" parameters that can be modified at will in the type of experiments suggested by our theory (Fig. 4) than in the conventional type. We will limit ourselves to pulses sequences characterized by:

1) identical pulses in energy and shape

2) predetermined statistical distribution of "pulse time of arrival"

3) predetermined statistical distribution of the phases of each pulse.

Our approach to the study of the influence of light coherence has a simple physical interpretation. The temporal distribution of pulses can be assimilated to a statistical distribution of giant photons. Instead of having a photon statistics of 10^{20} photons, we pick only a sample of 10 or 30, but of the same total energy, and average many experiments. The pseudo statistics will produce the same average number of ions as the real photon distribution, but it will take more than one ($\underline{N}$ 500) measurements to determine that value with a sufficient degree of confidence.

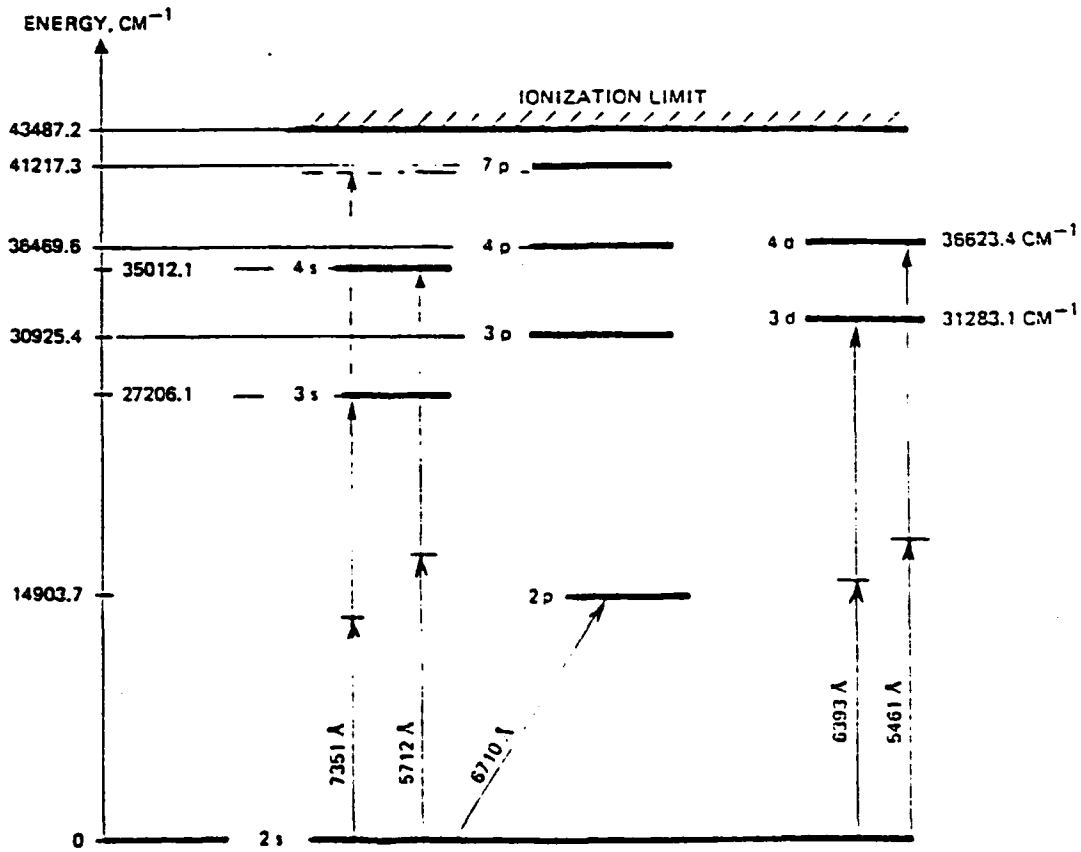


Figure 5. Energy level diagram of lithium (based on data from Ref. 14)

4. EXPERIMENTAL CONSIDERATIONS.

The source.

We have developed a source to the needs of this experiment. It consists in a synchronously pumped dye laser, followed by three stages of amplification pumped by a "Quantaray" frequency doubled Nd:YAG laser. The amplifiers are being pumped by 6 nsec pulses at a rep rate of 10 Hz, while the oscillator generates a train of pulses 12 nsec apart.

The major problems encountered in the development of the source were:

- pulsewidth control (oscillator) and pulse coherence;
- synchronisation (of the oscillator and amplifier);
- isolation between stages;
- pulse to pulse reproducibility.

1) Pulsewidth control. Pulse durations of up to 50 psec were obtained by a combination of 3 plate birefringent filter and two etalons in the cavity of the dye laser. The coherence of the source was checked by:

a) phase sensitive autocorrelations -peak to background ratio 8 to 1)

b) interference of a second pulse with the polarization created by a first pulse in lithium vapor[8]. By this experiment we also demonstrated our ability to produce a pulse sequence with the appropriate phase relationship for the coherent harmonic generation experiment.

c) self-induced transparency in lithium vapor, on the 2s - 2p transition.

2) Synchronization. (A circuit has been designed and built that divides a frequency of up to 500 MHz down to a few Hz with a jitter (fast and slow) of less than 1 nsec[9]. We modified, redesigned and rebuilt also the Q-switching circuit of the Quantaray laser, in order to reduce the jitter of the Q-switching command to less than a fraction of nsec.

3) Isolation between stages. We use a combination of

- spatial filters
- dispersive prisms
- 2 thin (0.3 mm) flowing dye cells of saturable absorber
- 1 color filter (saturable absorber).

4) Pulse to Pulse reproducibility.

Event though we attempt to saturate strongly the last amplifier stage, some pulse to pulse intensity fluctuation remain. We have therefore purchased a Smoke Signal Broadcasting microcomputer system Chieftain Model 811, which is capable of analysing and recording the data at 10 Hz of the source. All the hardware needed for this program (detector interfacing) has been constructed and assembled.

The atomic system.

We have demonstrated the tunability of our source to cover three transitions of lithium with two dyes: Rh6 (solvent: ethylene glycol for the oscillator and methanol for the amplifier and DCM (mixture of benzol alcohol and ethylene glycol as solvent). A simplified level diagram is shown in Fig. 5. By using readily available dyes, the following transitions can be studied:

- 2s-4s at 571.2 nm (Peak of the Rh6G emission). Resonant enhancement (laser induced resonance (section 3) may be provided through the 3d level and the blue (480 nm) argon laser line. Operation of

the laser oscillator-amplifier combination has been demonstrated at that particular wavelength.

- 2s - 3s transition at 7351 A. This transition requires a dye laser operating with nile blue or oxazine. This transition is of particular interest because "laser induced resonant enhancement" can be obtained by using the strong 1.06 m pulses from the amplifier pump laser (coupling through level 3d).

- 2s-3d transition at 6393 A. (Peak of the DCM emission). This transition would require the smallest power densities to reach the maximum efficiency (because of the proximity of the 2s-2p transition (single photon resonance at 671 nm, near 1/2 of the 2s-4s transition). The green light of the amplifier pump may be used for "laser induced resonant enhancement" (coupling through level 3s). The operation of the laser-oscillator amplifier has been demonstrated at the particular wavelength.

- 2s-4d transition at 546.1 nm (frequencies of a fluoresceine sodium dye). It is also of special interest because it is associated with a much smaller Stark shift than the other transitions. "Laser induced resonant enhancement can be obtained through the third harmonic of the 1.06 m Nd:YAG laser. Since this third harmonic is obtained by mixing the fundamental and second harmonic, the latter is still available to pump the 3 stage amplifier.

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